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INFLUENCE OF OPTICAL FIELD EMISSION ON THE NONLINEAR PHOTOELECTRIC EFFECT INDUCED BY ULTRASHORT LASER PULSES

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ABSTRACT

A decrease in the order of nonlinearity of photoelectron emission from a metal cathode was found experimentally with mode-locked laser pulses at sufficiently high intensities. This phenomenon is interpreted as due to the theoretically predicted appearance of optical field emission.

KIVONAT

Nagyintenzitásu mode-locking laser impulzusok hatására fémkatódból kilépő nemlineáris fotoelektron-áram fényintenzitás-függését vizsgáltuk. Elegendően nagy intenzitások esetén a nemlinearitás rendje erősen lecsökkent. A jelenség értelmezhető az elméletileg megjósolt optikai téremisszió felléptével.

РЕЗЮМЕ

Экспериментально обнаружено уменьшение порядка нелинейности фотоэлектронной эмиссии с металлического катода при использовании ультракоротких импульсов синхронизированного лазера достаточно высокой интенсивности. Это явление приписывается возникновению оптического туннельного эффекта, предсказанному теорией.

1. INTRODUCTION

A general formula was given by Keldysh [1] to describe the relation between the electron current j due to photoelectric effect or photoionization and the incident light intensity I /or field strength E / of high power laser beams interacting with solids or gases.

This formula is easily interpretable and applicable in two extreme cases:

a/ If for a given laser frequency ω, the field strength, is relatively low, the photoelectric current $j \sim (E^2)^n \sim I^n$ where $n = \left[\frac{A}{\hbar\omega} + 1\right]_{int}$, A being the work function or the ionization potential. Electron emission is then interpreted as due to the nonlinear photoelectric effect /NLP/ or photoionization /NLP/.

b/ If the field strength E is high enough one finds a limit at which $j \sim E^2 \exp\left(-\frac{\beta}{E}\right)$, where β is a constant. In this case the electron emission is due to the classical optical field emission /OFE/ at field strength E.

The quantitative criteria for NLP and OFE were defined by Keldysh as $\gamma >> 1$ and $\gamma << 1$, respectively, where $\gamma = \omega \frac{\sqrt{2mA}}{eE}$ / m is the mass, e the charge of the electron/.

Following Keldysh, Silin [2] showed that for metals there is a transition interval $n \geqslant \gamma \geqslant 1$, $\frac{\omega\sqrt{2mA}}{ne} \leqslant E \leqslant \frac{\omega\sqrt{2mA}}{e}$ between pure NLP and OFE, in which the photocurrent increases more slowly than is required by the dependence $j \sim I^n$.

The present work reports the experimental investigation of the NLP-OFE transition interval.

2. DISCUSSION OF THE EARLIER EXPERIMENTS

Relatively low values of E are sufficient for observing the NLP, and large number of reports have been published on its experimental and theoretical studies in gases and metals [1-21]. With giant pulses of $\tau \sim 10^{-8}$sec duration the observation of NLP at higher values of I is limited for metals by Richardson emission and sublimation, while for gases by avalanche ionization, saturation and resonance phenomena [5,10,11,19]. Thus, in order to achive intensities high enough for the transition to OFE while at the same time avoiding disturbing effects /except resonances/, ultrashort pulses of the order of $\tau \sim 10^{-11}$-10^{-12}sec are necessary [10] for both gases and metals.

The condition $\gamma \ll 1$ has already been obtained in gas breakdown experiments for sufficiently short τ [23], but the aim was, in this case, the investigation of the total ionization and not the relation $j = f(E)$.

With metals the problem is experimentally much simpler in many respects, as first the collection of ejected electrons is relatively easy, secondly, owing to the lower A, the value of γ is also lower. At the same time the disturbing effects /Richardson emission, sublimation etc./ can be suppressed by the use of ultrashort pulses and tangential incidence. Our former work on solids using mode-locked pulses [22] bore out these expectations, but owing to the $\gamma \gg 1$ condition the experiments were restricted to investigation of the NLP only.

3. DESCRIPTION OF THE PRESENT WORK

The transition interval $n \geq \gamma \geq 1$, as evaluated from the formula given by Silin [2], should start at $E \sim 10^{7.3}$V/cm and $I \sim 500$ GW/cm^2. which can be achieved with mode-locked Nd: glass laser pulses using a gold cathode. With the experimental arrangement shown in Fig. 1 the electron current j emitted from the Au metal cathode was measured as a function of the laser beam intensity I incident on the cathode.

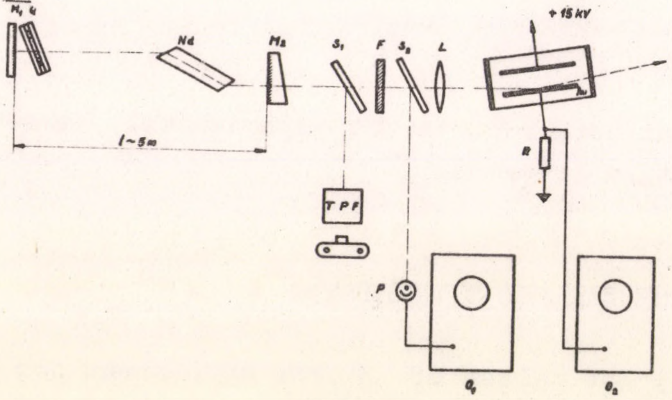

Fig. 1

The mode-locking laser arrangement consisted of two mirrors /M_1,M_2/ placed at a distance ℓ from each other, a passive Q-switch /Q/ and a neodymium glass rod /Nd/. The laser was built to meet the following requirements:

- maximum possible power density
- pure, picosecond mode-locking pulse train with large intervals between the pulse of a given train
- TEM_{00} mode
- relatively convenient reproducibility of trains.

The Brewster-ended Nd: glass rod was free from platinum, the mirrors were not damaged even at intensities as high as 20 GW/cm^2. The reflexion of the output mirror M_2 was 56%, while that of M_1 was ~100%. The Q-switch solution was placed into antireflexion cuvettes of various thickness /1mm, 500 μ, 40 μ/. Though the shortest pulses were obtained with the 40 μ cuvette [24,25], this was unsuitable for longer measuring runs, because the thin dye layer tended to lose its bleachability after a few shots, and thus it was employed here for control measurements only. For the actual measurements the 1mm cuvette was used instead. The spacing of the successive pulses of a train was 33 nsec, while the total length of pulse trains with $\ell \sim 5$m was about 400 nsec. Such pulse trains are convenient to handle electronically; moreover, the large value of ℓ permits a Frensel number of ~7 to be reached. By these means, and by delicate adjustment of the dye concentration, the pumping level and rate, TEM_{00} mode distribution could be achieved. The diameter of the laser beam was ~3mm, in spite of the 12mm diameter of the Nd rod, while its divergence was not more than $3 \cdot 10^{-4}$ rad. Under these conditions, with a shooting period of 2 minutes, the thermal lens effect could be minimized and the stability was found to be satisfactory.

The laser beam was reflected by the splitter S_1 to strike the TPF measuring setup, which served as a monitor of the picosecond structure of the pulse train. Its absolute intensity was determined from the calometric energy data and the pulse length calculated from the TPF data.

The part deflected by plate S_2 fell onto the fast biplanar photocell P /Type /, the output signal $V_L \sim I$ of which was photographed on a 1 GHz oscilloscope O_1.

The light intensity was varied by means of neutral filters F /NG Schott/, by the lens L, and by the fairly regular variation in the relative intensities of the pulses within the mode-locking pulse train. The linearity and transmission of the filters F were measured individually by a slight modification of the arrangement shown in Fig. 1, at the power density and wavelength used in the experiment. This measurement served simultaneously as a check on the electronic linearity.

The specially prepared 1 x 20 x 50 mm gold cathode was placed in a vacuum bulb at $\sim 10^{-8}$ torr. The laser beam struck this cathode at an angle of $\sim 5^{\circ}$.

The overall linearity of the measuring arrangement was checked by placing the monitor photocell in the path of the beam reflected from the gold cathode.

The electrons emerging from the cathode were collected on an anode kept at +15 kV potential. The output signal V_{NL} of the electron current j generated by a resistance R was photographed on the high sensitivity 100 MHz oscilloscope O_2 .

The light pulses emerging in the $6 \cdot 10^{-2} cm^2$ cross-section beam of TEM_{00} mode distribution had an average energy of 10^{-2} joule and a duration of $(5 \div 10) \cdot 10^{-12}$ sec. Thus the power density was ~ 16 GW/cm^2. The field strength on the target after focusing the beam diameter by a factor 2 was found to be $10^{6.8}$ V/cm ,this did not change on tangential incidence but considerably reduced the power density on the target and thereby prevented the occurrence of the above-mentioned background effects.

4. RESULTS

The amplitude of the associated pulse pairs V_L, V_{NL} of the pulse trains photographed on the oscilloscopes O_1 and O_2 were plotted in log - log coordinates to obtain the photocurrent versus light intensity curve shown in Fig. 2. For evaluation of the measured data the method described elsewhere was applied [22]. Owing to the more favourable laser operation, the asymmetry encountered in the earlier experiments [22] occurred apparently in a lower proportion of the shots.

The experimental points represent average values for 10 runs; the statistical errors are also indicated. The light intensity varied from about 6,5 to 66GW/cm^2, which corresponds to field strengths of from $10^{6.3}$ to $10^{6.8}$V/cm . The nonlinear photocurrent varied between $0.5\text{-}5 \cdot 10^2$ A/cm^2 . In the calculation of current density the real dura-

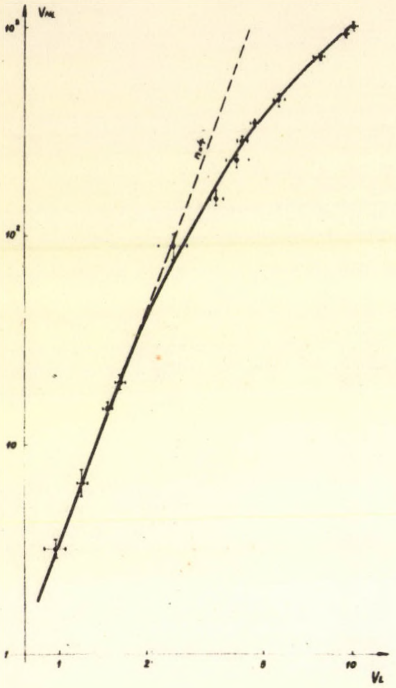

Fig. 2

tion of the electron pulse /i.e. the time during which the light pulse runs along the surface of the cathode, which is longer than the duration of the light pulse/ must be taken into account.

The shapes of the V_{NL} trains belonging to the V_L monitor trains of approximately similar length /half values/ at different intensity intervals can be seen on the Polaroids of Fig. 3; frames on the left are the V_L trains, those on the right the corresponding V_{NL} trains. It can be seen that the nonlinearity decreases in the order a, b, c, from low to higher intensities, while the half-width of the train inses and approaches that of the monitor train.

It is apparent from the curve in Fig. 2 that the formula $j = \alpha(E^2)^n$ with $n = 4$ holds‡ up to a field strength of $\sim 10^{6.6}$ V/cm.

For α_{exp} we have $10^{-67\pm3}\left[\left(\frac{A}{cm^2}\right)\left(\frac{m}{V}\right)^8\right]$, as compared with $\alpha_{theor} = 10^{-69}\left[\left(\frac{A}{cm^2}\right)\left(\frac{m}{V}\right)^8\right]$ predicted by the theory [13] for $A=4.8$ eV; $\omega=2\cdot10^{15}$Hz; $n=4$. Above this field strength the slope of the curve gradually reduces down to $n \sim 1.2$. Comparing this result with the theoretical value of $10^{7.3}$V/cm, it seems that the transition interval starts at a lower field strength than is predicted by the theory.

To prove that the effect obtained is a real decrease in the order of the nonlinearity of the $j \sim f(E)$ dependence, the following controls were performed:

1. The ratio of the density of the conduction electrons and the number of electrons emitted during a single pulse /> 10^5/ ensures that a saturation effect similar to that observed in photoionization of gases [5,11] cannot occur.

2. The absence of space-charge and nonlinearity of the electronic system were controlled by the following measurements, besides the methods mentioned above:

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‡A slight increase of the slope calculated in [26] is not excluded, considering the value of errors given here.

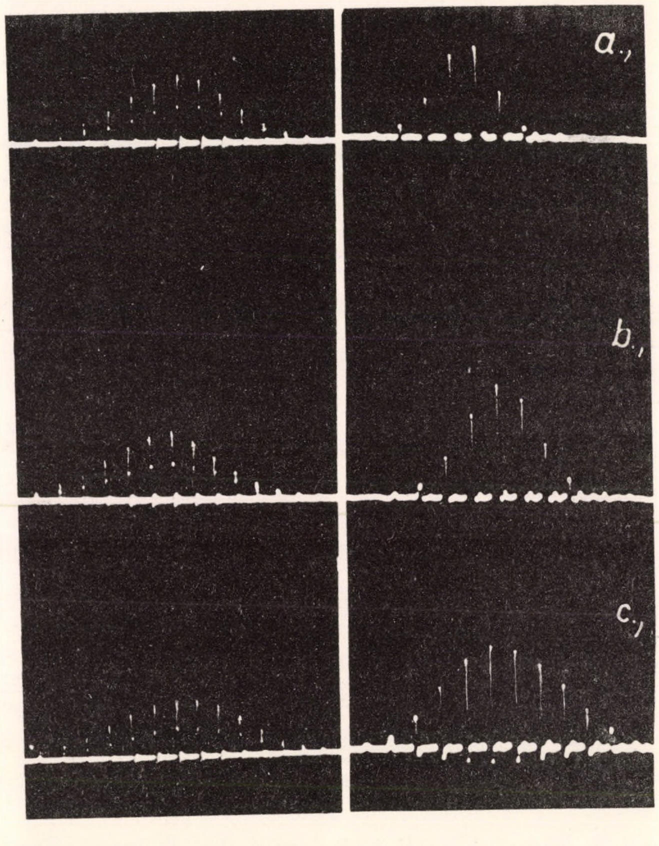

Fig. 3

- The effect was independent from the accelerating potential in the 3 kV - 25 kV range. The influence of space-charge was experienced under 3kV only, so that the 15kV potential used during the experiment is well above the space-charge threshold.

- The linearity of the electronic system is also proved by the fact that if constant photon density is maintained as the beam is widened /by increasing the number of photons in the beam/, the signal remains proportional to the beam cross-section.

3.The effect occurred independently of the method /neutral filters or lens/ by which the light power density was changed.

5. CONCLUSIONS

According to our experimental results there is a decrease in the order of the nonlinear relationship between the photocurrent and light intensity as the laser power density is increased. A similar decrease is predicted by the theory [2] for the NLP - OFE transition. However, for the critical field strength of this transition a somewhat lower value was found then is given by the theory. To decide whether the idealized theoretical model or the estimation of the absolute value of intensity by TPF method is responsible for this difference, further theoretical studies [27] and experiments with single ultrashort pulses and direct measurement of pulse duration are in progress.

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